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TREATMENT OF OIL SPILLS WITH NATURAL SORBENTS: A REVIEW

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Abstract: **One of the most available high-energy-density fuels is fossil oils. As a result, people from every corner of the globe have sought fossil oils. They are usually sourced away from internationally recognized sites. Therefore, long-distance transport is required during which oil leakages become a problem. Oil spills and chemical leaks occur regularly as oil production and marine traffic expand. The release of effluents from the oil and gas industry has posed a severe environmental problem as its disposal has not been effectively curbed. Resolutions have been made and are still being made to solve the problem. With this, works from several researchers have shown the feasibility and working progress of applying separation techniques to obtain valuable products from the effluents. One of the areas that have been majorly studied is the adsorption of oil using inorganic adsorbents. Noting that most of the inorganic adsorbents are expensive, the use of agricultural biomass as precursors for adsorbents is now being considered, as they are relatively cheaper means. This review reports the impact of oil spills on the environment and highlighted the remediation of oil spills with organic adsorbents.**

Keywords: **Environment; Fossil oil; natural adsorbents; oil spill; water treatment.**

1. INTRODUCTION

An oil spill occurs as oils flow out from the ground of oceans or unexpected oil spillage, from storing, managing, shipping, upstream activities and scheduled maintenance operations, entering the aquatic ecosystem. Oil spills have been seen on a regular basis for nearly thirty years, which is one of the primary sources of pollution in water bodies. In addition, the levels of oil transport and weathering also rise due to factors such as salinity, temperature and tides [1]. Over several decades, some 1.5 million tons of crude oil have spilt in the Niger Delta, which has not been completely cleared up and resulted in wastelands. [2] has estimated that the petroleum industry has raised its rate of oil spillage from 1976 to 2008 to around 80%. Oloibiri, a town located in the Niger Delta area of Nigeria, was found to have oil oozing out of the ground to a minimal extent before finding the oil in significant quantities in 1956. However, the growth of petroleum business activities has intensified oil spills and the consequent environmental degradation [2] .

Between 1970 and 2010, approximately 5.71 million tons of oil spilt due to tanker accidents. The physical and chemical properties of the oil spill and oil slick had a major impact on marine life, seas or water supply system, tourism, and recreational facilities. Climate, rate of spreading of oil on the surface of the water, movement in the marine environment, vaporization into the air, and emulsification of water and oil all play a role in the formation of oil slick after a spill [3]. The Deepwater Horizon oil spill that occurred in 2010 has brought approximately 4.9 million barrels of petroleum into the Gulf of Mexico. The oil spill harmed marine life, plants and the inhabitants on the coast of the Gulf of Mexico. Various remedial techniques, including chemical dispersing agents, mechanical booms and skimmers, have been used to counter the risks of the leaked oil on the setting. To mitigate the effects of the spilt oil, more than 2.1 million gallons of the dispersing agents were used. The presumed toxicity of the dispersant formulations was nevertheless causing human and ecological toxicity problems [4].

Environmental degradation occurs during oil and gas processing when oil is transported to the atmosphere by transit, refining, and use. Total global oil emissions from the transport of petroleum hydrocarbons alone has been estimated to be

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up to 10 million metric tons. The concentration of oil-polluted water resulting from industrial operations could rise as high as 40,000 mg/l [5]. With this figure, quick, efficient, and environmentally friendly techniques need to be developed to combat oil spills, considering the effects the incidence can have on the environment.

2. EFFECT OF OIL SPILLS ON THE ENVIRONMENT

Petroleum's environmental consequences are primarily harmful and detrimental to almost all lifestyles. Oil emissions can be hazardous and poisonous to the human body in the air and in the sea. A significant issue for the oil sector is separating oil-polluted water from oil without affecting the environment. Pollution of water resulting from such incidents has resulted in significant environmental and ecological challenges and has been a major threat to the climate, human health and national health [6] . Spilt oil in the marine habitat can have severe economic repercussions on both shoreline activity and the exploitation of ocean resources [7]. Thus, there is a need for treatment of these effluents released into the environment for health concerns and the maximization of all resources.

During oil and gas processing, environmental degradation occurs when oil is transported to the atmosphere by transit, refining, and use [8]. Total global oil emissions from the transport of petroleum hydrocarbons alone has been estimated to be up to 10 million metric tons. The concentration of oil-polluted water resulting from industrial operations could rise as high as 40,000 mg/l [5]. During crude oil production, the water provided with crude oil also has to be handled. It should be isolated and discarded in a way which is not inconsistent with ecological sustainability [9]. The disposal methods of oil-polluted water were extensively analyzed and discussed by preliminary researchers [10].

Conventional strategies for oil spill cleanup involve the use of sorbent materials; adsorption is extensively used for remediation purposes to remove a large range of contaminants such as metals and organic compounds [11]. Carbon-based materials are the most commonly used sorbents for adsorbing dissolved contaminants at low concentration. Adsorption onto carbonaceous materials including activated carbon (AC) has been extensively studied for the removal of a wide variety of pollutants from wastewater and sediments [12,13]. The excellent adsorption efficiency of carbonized bio-materials is primarily attributed to their large surface area and their widespread use ensures that carbon - based materials are commonly used for this purpose [14].

3. METHOD OF REMOVING OIL SPILL

Numerous tactics for remediation are followed when oil spills occur, in order to reduce their environmental effect. In-situ burning, mechanical trapping and recovery, and chemical dispersing agents are some of the most widely used oil spillage responses. Oil absorbents have recently become the target of many researchers as a remediation alternative, making it possible for the spilt oil to be recovered [15-17]. However, it is essential to note that free oil and emulsified oil are two different types of petroleum-based effluent. As it can be isolated by gravity and skimmed off, free oil is simple to extract. Consequently, because emulsified oil has better stability in the aqueous medium, it is more tasking to extract [18].

Before knowing the beneficial aspect of biomaterials in oil spill management, three methods are widely used for the recovery of oil from the surface of the environment (Fig. 1). Different methods for remediating oil spill are grouped as physical/mechanical, chemical, or biological.

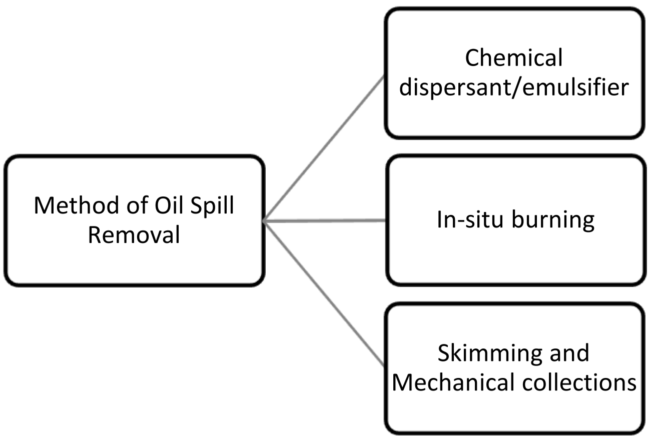

Figure 1: Methods of Oil Spill Removal

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The mechanical approach is the most labour-intensive method for oil recovery, involving the oil taken up by a sorbent. This technique utilizes a net-like structure containing sorbent material that encircles the spilt oil and gradually shifts to capture the oil on the surface [19,20]. Gupta and Tai (2016) [21] found that in conjunction with being environmentally friendly, reusable, low density, and having excellent buoyancy properties, the sorbent is durable and chemically stable for the handling of spilt oil. Physical and chemical methods are not employed in any type of spilled oil occurrence due to the risk of polluting the atmosphere, high cost, and limited adsorption capability. Instead, biodegradable, low-cost biomaterials are commonly used for removing spilled oil. Biomaterials come in various shapes and sizes, including kapok, cotton, rice straw, barley, oats, rice husk, wheat, and many others. They are readily available, cost-effective, biodegradable, and non-toxic, making them suitable as sorbents in oil spill response [22].

Research has indicated that organic matter can be used as sorbent, surfactant and separator in remediating oil spillage, but it can interfere with the natural ecosystem. Thus, there is a need for environmentally friendly, harmless, and inexpensive organic material to effectively remove and recover oil [22]. Biomaterials that function as sorbents for small-scale oil spill occurrences are commonly used. An excellent oil sorbent material will have a high adsorption capacity, hydrophobicity and oleophilic, biodegradable, and reusable. The use of biomaterials stand out from all other strategies and processes because of their unique properties [23,24]. Natural sorbents remove a lot of oil and preserve the state of the environment while recovering the oil. Research has shown that some plants have a strongly attract oil. That is attributed to the prevalence of wax on the species of plant. As much as these natural sorbents have high adsorption potential, they also have some drawbacks, like the potency to take up water during the oil cleanup process in an aqueous phase. According to research, a few natural sorbents have shown higher adsorption capacity than others, like raw cotton and kapok fibre.

After the cellulose from a natural fibre has been modified by acetylation, the higher oleophilic quality of Kapok fibre (20–50 g/g) or cotton allows for a higher affinity for oil, which aids the biomaterial inadequately adsorbing spilt oil [25,26]. Raw cotton can also extract oil from the surface, but the cotton gains more durability and biodegradability after modification by acetylation [22]. Kapok is a fibrous material that has a high capacity to absorb oil. It has a high potential for oil absorption because of the porosity of its structure [27]. To boost the oil adsorption capacity, many surface treatment methods are employed, like Esterification and acetylation. The high amount of hydroxyl groups in kapok fibre's original structure is responsible for its low oil sorption potential. This structure, on the other hand, can be altered to produce a surface that is highly oleophilic and hydrophobic [22].

Annunciado et al. (2005) [28] investigated the use of various vegetable fibres, namely mixed leaves residues, mixed sawdust, sisal (Agave sisalana), coir fibre (Cocos nucifera), sponge-gourd (Luffa cylindrica) and silk-floss as sorbent materials of crude oil. Sorption tests with crude oil were conducted in deionized and marine water media, with and without agitation. Water uptake by the fibers was checked by tests in dry conditions and distillation of the impregnated sorbent. The silk-floss fibre showed a very high degree of hydrophobicity and oil sorption capacity of approximately 85 g oil/g sorbent (in 24 hours). Specific gravity measurements and buoyancy tests were also used to evaluate the suitability of these fibres for the intended application. In all, irrespective of the sorption conditions, the 24-h oil sorption of the silk floss reached approximately 85 g oil/g sorbent. This sorption capacity is much higher than those reported in the literature for other vegetable fibres. Witka-Jezewska et al. (2003) [29] showed sorption values from different authors and a maximum of 40 g oil/g sorbent for unscoured cotton. Saito et al. (2003) [30] reported a maximum of 16.5 g oil/g sorbent for Sugi Bark. Barley straw, sawdust, cross-link cellulose, and oat straw are also utilized to effectively take up oil in the oil cleanup process. Sorbents used in the oil cleanup process must be oleophilic and hydrophobic, specific, high recovery level, inexpensive, environmentally safe, pollutant-free, and biodegradability [22].

Organic Adsorbents

Innovative environmental friendly, and sustainable technologies for removing spilt oil are urgently needed. Activated carbon, carbon nanotubes, organic metal frameworks [31], and minerals have also been tested for oil-water separation [32,33]. Agricultural wastes can also be utilized as adsorbents for oil-water separation, thus lowering the cost of the adsorption and reducing the environmental effect of producing adsorbents. A successfully prepared adsorbent must be low-cost and have adequate adsorption properties and the potential for reuse [34].

Rice husk is a biodegradable matter processed in a substantial majority, primarily released as a by-product from the rice milling and agro-based biomass industry [35]. It is expected that for every ton of rice grown, 0.23 tons of rice husk is

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obtained simultaneously, resulting in approximately 1 million tonnes of rice husk being obtained annually worldwide [36]. RH is mostly chemically identical to many widely used organic fibres, comprising silica, hemicelluloses, lignin and cellulose, with silica as the principal constituent of ash. The rice husk ash (RHA) is collected, having broken down the organic matter by burning [37]. Among other agricultural wastes, RHA is rich in silica content, with about 90 percent to 98 percent of it and a low fraction of metallic contaminants completely combusted [38,39] (Ahmed et al., 2008; Real et al., 2010). It is crucial that the silica in RHA is amorphous and possesses a large surface area [40,41].

The preparation of silica nanoparticles can be done in a variety of ways. Adam et al. (2011) [42] used the sol-gel method to make spherical nanoparticles from rice husk. The silica particles produced were discovered to be agglomerates with average dimensions ranging from 15 to 91 nanometres. Jal et al. (2004) [43] used the precipitation technique to make silica nanoparticles, which had a particle size of 50 nanometres in dimension. The sol–gel procedure [44] is, however, the widely used approach for the synthesis of silica nanoparticles. It entails both condensation and hydrolysis reactions simultaneously [45]. Jyoti et al. (2021) [46] also used Leaching and co-precipitation to synthesize amorphous nano silica from rice husk.

Rice straw is another agricultural waste, which is obtained after harvesting rice grains. It is usually fed to ruminants and can be applied as an oil sorbent. According to some studies, its use as an adsorbent can be attributed to the prevalence of cellulose in its cellular membranes. [47] Gongtested the modification of rice straw with phosphoric acid and citric acid without any alkali pre-treatment [48,49] as possible adsorbents for dye removal from water. Chakraborty et al. ([50] used sodium hydroxide to modify treat rice husk as an adsorbent for the adsorption of crystal violet dye. They discovered that the monolayer adsorption power increased to 44.87 mg/g. They found that treating rice husk with sodium hydroxide breaks the covalent bond between lignocellulose materials, hydrolyzes hemicellulose, and depolymerizes lignin, thereby increasing adsorption potential.

Bio-based materials have recently attracted a lot of attention in industrial applications [51]. Table 1 shows the most popular natural fibres manufactured around the world utilized as adsorbents. Natural fibres are gotten from plants and animals, and are composed of cellulose, hemicelluloses, lignin, extractives, and inorganics. They are also hydrophilic given the prevalence of highly polarised hydroxyl groups in the lignocellulose. As a result, the hydrophobic polymer matrices are simply incompatible with these fibres. It is critical to study the nature of these fibres before beginning certain treatments or alterations [52]. Several researchers had focused on cellulose because of its unique properties, such as hydrophilicity, easy processing, non-toxicity, and low solubility in a good number of solvents due to the presence of hydrogen bonds and crystalline nature [53]. Since the hydroxyl groups in these glucose units are most reactive, they are essential in modifying the fibres.

Table 1: Natural Fibres utilized as Adsorbents

Type of Adsorbent	Percentage of oil removal (%)	References
Wool	88-93	[54]
Bark	95	[55]
Rice Husk	78-94	[56]
Banana	85	[57]
Kapok	75	[58]
Oil palm leaves	90	[59]
Sugarcane bagasse	99	[60]

Chemical modification of organic adsorbents

Owing to its favourable results, relatively low cost and convenience compared to carbonization, chemical treatment is seen as a possible application for increased oil efficiency. In past efforts to increase the performance of oil removal through chemical modifications, several methods were applied to natural sorbents. Solvent treatment with acid and alkali, surfactant treatment, salt treatment, Esterification, and acetylation are used in modifying natural fibres [5]. The sustainability and adsorption capacity of biosorbents are improved by chemical treatment [61] (Demirbas, 2008). [62] confirmed that cellulose could not serve as an adsorbent without being modified. In this regard, Wang et al. (2012) [27]

investigated the use of hydrochloric acid (HCl) to treat kapok fibre as as a natural sorbent. It was discovered that wax was removed by adsorption, and the number of cellulose hydroxyl groups in the kapok fibre walls increased on account of the treatment. In the analysis, the HCl-treated kapok fibre's oil sorption capacity was found to be between 11% and 27% more than the untreated kapok fibre.

Another research was carried out by [63] Wang et al. (2018) to analyze the effectiveness of modified rice husks for oil spill cleanup. In this study, the most effective treatment technique for the development of cellulose fibres from rice husk was selected by comparing different biological treatments. *A. flavus* treated rice husks had the highest cellulose percentage and were chosen to be acetylated. The acetylation process was found to have further increased its oil adsorbing capacity and hydrophobicity. The acetylated fibre had shown an oil adsorption capacity of 20 g/g. [32] Lin et al. (2014) tested carbonized rice husk treated with polyethlyenimine (PEI-CRH) for application in the adsorption of oil from water. PEI-CRH was made by impregnating the carbonized rice husk (CRH) with polyethlyenimine (PEI). PEI-CRH had a significantly better oil-water adsorption efficiency (about 10–50 times) than pure CRH. PEI-CRH was discovered to be capable of absorbing up to half its mass in oil.

In the esterification process, the ester functional groups (O–C=O group) are bound to cellulose hydroxyl groups through precursor condensation in the esterification process [64]. Based on the type of reagent applied, Esterification may cause fibre damage. [65] Hubbe et al. (2015) explained using alkydes, carboxylic acid, acid chloride or alketonedimers, forming ester bonds with the cellulose hydroxyl groups. Modification by Esterification has shown the capability of improving the cellulose's hydrophobicity [66,67], which could help with adsorbing oil. According to Asadpour et al. (2016) [68], the viscosity of the oil plays a significant role in the sorption process. The acetylated oil palm empty fruit bunch (OPEFB) fibres adsorb oil with low viscosity way quicker than highly viscous oil in this study.

Alkaline treatment is regarded as a cost-effective therapy, as it aids in the removal of wax and lignin from the fibre's surface. Furthermore, it boosts crystallization potency, facilitates interfacial interaction, and improves fibre functionality all at the same time. As a result, it aids in enhancing the mechanical properties of the fibres [69]. Alkali treatment is primarily applied to eliminate lignin, pectin, wax and natural oils that coat the fibre's external surface, improving its top layer and mechanical performance with application to polymers [70]. This was evident when Li et al. (2015) [71] observed an enhancement of 50% in the mechanical properties of bamboo fibre that had undergone alkali treatment. Fathy et al. (2013) [72] also investigated the efficacy of alkali-acid modification in improving rice straw (RS) adsorption efficiency in removing a basic dye. They discovered that pre-treating the rice straw adsorbent with an alkali acid improved its adsorption ability. The adsorption capacities of unmodified and modified RS samples with NaOH-1M citric acid differed from 32.6 to 131.5 mg/g.

Combination of adsorbents

An experiment was conducted by Malekmohammadi et al. (2016) [73] to study and compare silica, activated carbon, and zeolite adsorbents in the removal of ammonium, iron, COD, turbidity and phosphate pollutants and to investigate the effect of discharge on the removal of contaminants (Table 2).

Table 2: Final concentration of pollutants after passing through silica, activated carbon and zeolite columns

Pollutant	The initial concentration of pollutant	The concentration after silica	The concentration after zeolite	The concentration after zeolite	The maximum permissible level in drinking water
Ammonium (mg/L)	5.5	4.7	1.5	3.5	1.5 mg/L
Iron (mg/L)	0.55	0.1	0.5	0.35	0.3 mg/L
Turbidity (NTU)	100	8.1	-	9.7	5 Ntu
Phosphate (mg/L)	4	2.8	1.2	2.5	-
COD (mg/L)	200	70	180	21	0

Source: [73]

The table above shows the data obtained on analysing the pollutants present in a water sample. In drinking water, these levels were compared with the overall allowable volume. As shown, silica has a high potential to extract iron and turbidity and can reduce the concentration of iron to the permissible degree. Ammonium, phosphorus, and COD are reduced by silica but do not exceed the acceptable quantity. Zeolite has a fantastic potential to remove ammonium and phosphate and can reduce the permitted amount of ammonium. But, with the replacement of iron and COD, its capacity is marginal. Activated carbon reduces turbidity and COD significantly and decreases other contaminants, although it does not exceed the highest allowable drinking water amount.

The separation yield was more than the other adsorbents for iron contaminants using silica adsorbents. The removal yield obtained from zeolite was marginal, however. In removing ammonium and phosphate, the zeolite column has the best quality. Also, in the activated carbon column, eliminating these two pollutants is safer than silica. The COD removal yield using activated carbon is higher than silica, although its zeolite removal yield is insignificant. Silica has the best potential to dissolve iron and turbidity, while zeolite can remove ammonium and phosphate efficiently. In addition, the safest adsorbent for COD and turbidity reduction is activated carbon. This suggests that each adsorbent may eliminate a particular form of pollutant. Accordingly, it is planned to combine adsorbents for the elimination of contaminants. The previously observed trend from the study carried out already shows that are likely lapses for each of the adsorbents. At the same time, they are used separately, which now encourages a combination of the adsorbents. Dawodu et al. (2020) [74] also looked into using a feldspar-banana peel biochar composite as a low-cost, innovative adsorbent for removing crude oil from an aqueous medium. The prepared adsorbent brought forth a heterogeneous surface that exhibited improved hydrophobicity and better adsorptive capacity for the adsorption of crude oil.

4. CONCLUSION

This review paper has brought forth several works that have been done to remediate oil spills. Sorbents from biomaterials have been depicted as environmentally friendly, biodegradable and low-cost, making them advantageous to the environment. However, they need to undergo modification to increase adsorptive capacities through their hydrophobicity, oleophilic capacity, porosity and other factors. Moreover, there is a need for more researches on combining these adsorbents from organic matter, as all adsorbents are not sufficient for their respective applications.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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